Search for X-ray Induced Acceleration of the Decay of the 31-yr Isomer of ^{178}Hf Using Synchrotron Radiation

The intense flux of high-energy x-rays from the Advanced Photon Source has been used to verify claims in the literature of an extraordinary cross section for triggered decay of the 31-yr ^{178}Hf isomer, induced by x-ray bombardment. The detailed analysis of the results sets an upper limit for the cross section over a wide range of incident photon energy (from 8 to 100 keV) consistent with nuclear physics estimates for the process and orders of magnitude below the previous work. The prospect for practical use of nuclear isomers as energy-storage devices, triggered on demand, is still speculation.

Releasing the energy stored in an isomeric nuclear state with an atomic trigger is an attractive speculation: the energy gain is on the order of the ratio of nuclear/atomic energies—MeV/keV. (Nuclear isomers are loosely defined as excited nuclear states with lifetimes longer than 10^{-9} s.) The landscape of nuclear isomers includes examples with lifetimes on the order of years. These isomers may be thought of as energy-storage materials with an energy release 10^4 to 10^5 times that of chemical energies. The energy release can be either slow or rapid. Nuclear isomers therefore represent an opportunity for a standalone energy source if suitable schemes for trigger and control of the energy release can be identified. Potential applications include space drive, as well as very bright γ-ray sources [1].

The excited states of ^{178}Hf include a well-known example of a nuclear isomer with excitation energy E_x = 2.447 MeV, spin-parity $J^\pi = 16^+$, and half-life $t_{1/2}$ = 31 yr (Fig. 1). The decay scheme of this isomer to the nuclear ground state is well known. The 2.447-MeV isomeric state decays slowly ($t_{1/2}$ = 31 yr) to the nearby state at 2.433 MeV. The 13^- state loses energy in a rapid (t ~ 10^{-12} s) γ-ray cascade ending at the 8^- rotational band head, which in turn decays via the ground-state rotational band cascade. The γ-ray cascade is delayed at the 8^- state at 1.147 MeV, since the 8^- state is also isomeric, with $t_{1/2}$ = 4 s. Very scarce quantities of the 16^+, 31-yr isomer are available for research (~ 10^{15} atoms).

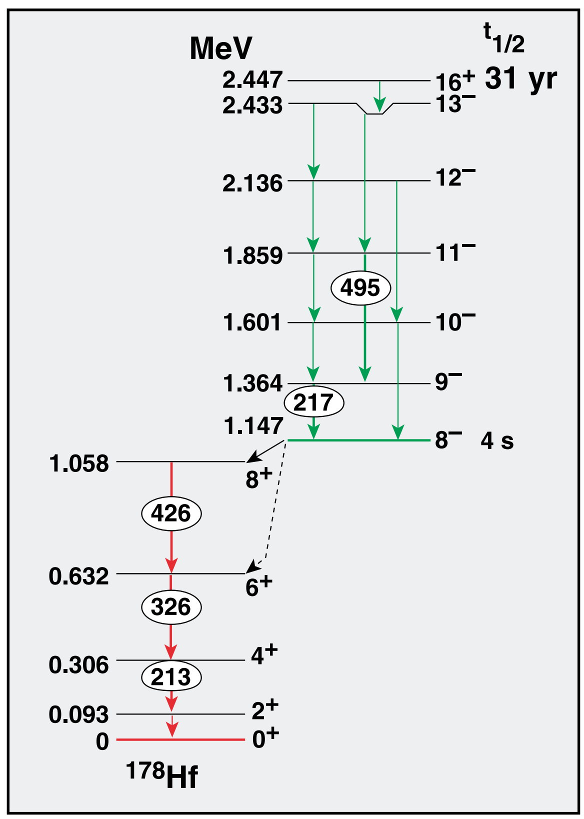

FIG. 1. Nuclear energy level diagram showing the decay of the 31-yr ^{178}Hf isomer. Transition energies (in white ovals) are in keV and level energies in MeV. Transitions reported as enhanced are indicated.

Triggered decay of the ^{178}Hf isomer induced by x-rays delivered by a dental x-ray machine has been reported [2] and affirmed [3]. Enhancements ε amounting to ~1–2% in the isomer decay rate ($dN/dt = -(1 + \varepsilon)N/\tau$ have been reported for various γ-rays in the cascade (distinguished by red and green vertical lines in Fig. 1). The reported integrated cross section for triggering the decay is 10^{-21} cm^2 keV, so large as to demand new physics [4]; it is at least 10^7 times expectations from nuclear physics. The large cross section has been viewed with skepticism; nevertheless, the persistent experimental results suggest an independent investigation because of their significance for the many applications for energy storage and its controlled release.

The intense photon flux available at the DOE Advanced Photon Source (APS) offers the opportunity for orders of magnitude improvement in experimental sensitivity. A team of scientists from Argonne, Los Alamos, and Lawrence Livermore National Laboratories formed a collaboration; made an experimental design that took advantage of the high APS flux and that also featured low systematic errors; irradiated samples of HfO_2, which included ~10^{15} atoms of isomeric 31-yr ^{178}Hf; and searched for signals characteristic of an enhanced (speeded up) isomer decay. The sample was fabricated at Los Alamos National Laboratory using material chemically extracted from the LANSCE/LAMPF target/beam stop. (The reaction mechanism was spallation induced by the 800-MeV proton beam.) Irradiation with the intense "white" beam of photons was chosen since the "trigger" photon energy is uncertain. The irradiation was done at the SRI-CAT beamline 1-ID [5] in March 2001. The undulator was operated with maximum taper (5 mm) in the gap and two average gap settings: 15 mm and 20 mm. This arrangement generated a smooth "white" photon flux peaking at ≈ 2 × 10^{15} photons/keV-s at E_{ph} ≈ 16 keV and extending in energy to well over 100 keV (Fig. 2). The beam power on sample was limited to 210 W to avoid damage to the sample encapsulation, a water-cooled Al assembly with an entrance window 0.15 mm thick. In addition, the sample assembly was enclosed in a containment vessel during irradiation. The entire system met APS safety

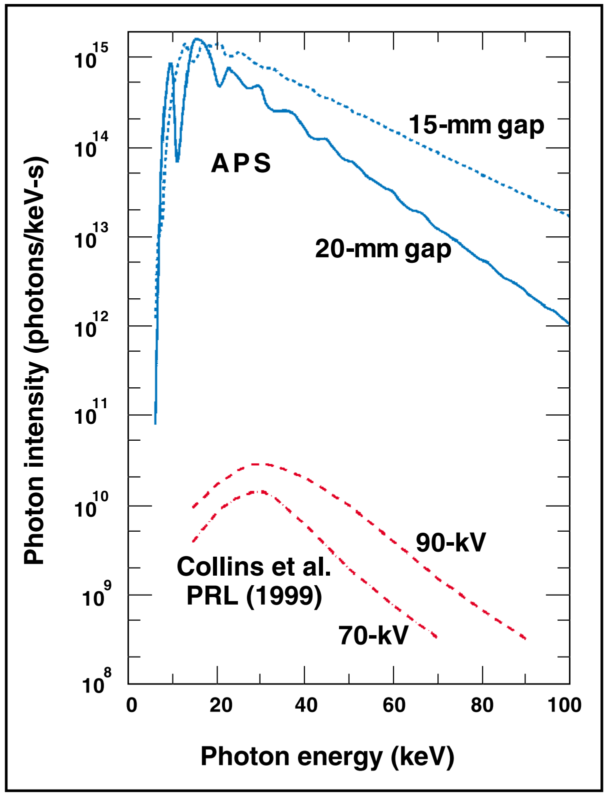

FIG. 2. The calculated photon intensity from the Advanced Photon Source, incident on the 2-mm-diameter area of the target material in the present experiment, for the two tapered settings of the undulator gap that were used. Photon intensities given in previous experiments [2] for the 70- and 90-kV settings of the dental x-ray machine on a 1-cm diameter target are also shown.

requirements. The 1-ID white beam was mechanically chopped to form a pulse train of 11-s beam-on and 22-s beam-off during the irradiation intervals, ≈ 8 h for each of the three samples. Precision γ-ray spectrometers based on Ge detectors with energy resolution characterized by FWHM ≈ 1.0 keV at E_{ph} = 300 keV were used to count the sample during the irradiation cycles. Individual γ-rays characteristic of the ^{178}Hf isomer decay were easily identified in the γ-ray spectra, along with beam-induced fluorescent Hf K x-rays. These x-rays were used to monitor beam incident on the sample; the measured value agreed within a factor of two with the calculated beam flux. The experimental signal of triggered isomer decay is an increase in the yield of characteristic ^{178}Hf γ-rays, above the background decay rate of the 31-yr isomer. The experimental design gave the experimental team two chances to observe any

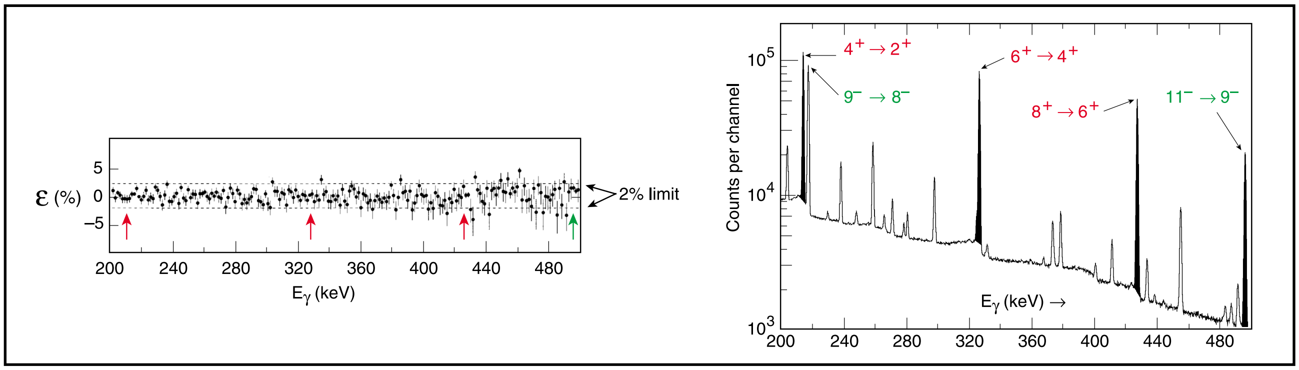

FIG. 3. A partial γ-ray spectrum of isomeric 31-yr ^{178}Hf. Photopeaks for the transitions at 213, 217, 326, 426, and 495 keV are filled in for the previously reported enhanced transitions. The spectrum is an accumulation of ~22-s counting periods, immediately following the 11-s irradiations of the R1 sample. The average undulator gap for these data was 15 mm, and the data were accumulated over 8.5 hr. The points with error bars show the difference spectrum between the first half and the second half of the 22-s counting interval, in percent. This difference, with the points summed over an energy interval corresponding to the detector resolution, reflects any triggered excess in de-excitation through the 4-s isomer. The dashed lines indicate 2% limits in the difference.

increase in isomer depopulation: during the 11-s beam-on sample intervals and during the 22-s beam-off interval. Analysis of the spectra obtained in the beam-off interval is especially attractive because (1) there is no background from scattered beam photons, and (2) a fraction of the decay γ-rays are caught and held up in the 4-s, 8⁻ level midway down the decay chain and therefore exhibit a characteristic 4-s half-life (Fig. 1). Counts due to isomer triggering will be in excess of the expected counts from the radioactive 31-yr isomer at the beginning of the 22-s counting interval and will decay away with a 4-s half-life during the 22-s counting interval, leaving the background of counts from the 31-yr isomer.

A straightforward procedure for isolating decays due to triggering that cascade through the 4-s isomer is to divide the 22-s counting interval into two equal parts; integrate the γ-ray spectra over time of the corresponding halves, C(1) and C(2); form the difference spectrum C(1) − C(2); and identify net counts correlated with the characteristic of the ^{178}Hf isomer decay γ-rays. Several HfO_2 samples of different thickness were irradiated during the experimental beam time. Our best individual result came from the HfO_2 sample R1 containing 7.3×10^{14} isomeric atoms. The result is shown in the left portion of Fig. 3, which expresses the count rate difference as [C(1) − C(2)]/C(2), in percent. The right portion of Fig. 3 illustrates the γ-ray spectrum associated with the decay of isomeric ^{178}Hf [2]. Clearly there is no net positive enhancement for the characteristic γ-rays identified earlier (labeled in Fig. 1); instead the decay rate is constant within 2%. The

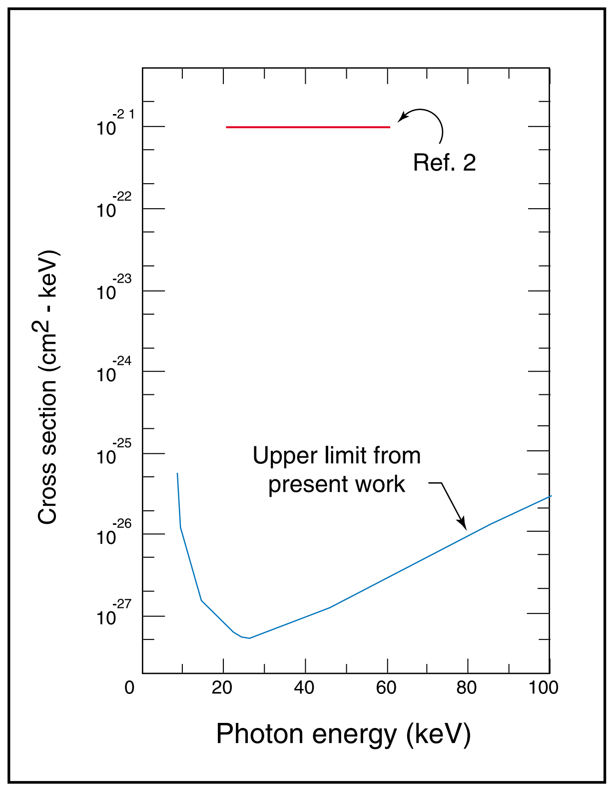

FIG. 4. Upper limit of the cross section for photon-induced de-excitation of the 31-yr ^{178}Hf isomer through the 4-s, 8⁻ isomer based on the measurements reported here. The value for this cross section reported in Ref. 2 is also shown. Other values reported by this group in the sources cited in Ref. 3 range from 2×10^{-22} to 2×10^{-21} cm^2 keV. The present limits are substantially below all previously reported values over the entire energy range.

number of triggered events is given by the product of the cross section for triggering σ, with the incident photon flux and the number of isomeric atoms per unit area, ϕ N/A, well-characterized quantities. Our experimental upper limit to the integrated cross section for isomer depopulation σ_{int} (after correction for self-absorption in the HfO_2 sample) is illustrated in Fig. 4. The limit is less than 2×10^{-27} cm^2 keV for incident photon energies between 20 and 60 keV, more than five orders of magnitude below the previous positive reports. At the lower incident photon energies of 15, 10, and 8 keV, upper limits are less than 2×10^{-27}, 10^{-26}, and 5×10^{-26} cm^2 keV, respectively. (At the lowest energies, self absorption of the photon beam in the Hf sample becomes important.) The present upper limit is clearly discrepant with earlier work over the entire energy range of incident photons. Finally, the limit discussed here is for triggered decay cascading through the 8^- state; if the triggered decay mode bypasses the 8^- state, the limits are a factor of 10 higher. More details of this experiment and the results can be found in Ref. 6.

The goal of the experiment was to verify claims in the literature of an extraordinary cross section for triggered decay of the 31-yr ^{178}Hf isomer, induced by x-ray bombardment. Our result sets an upper limit for the cross section over a wide range of incident photon energy (from 8 to 100 keV) consistent with nuclear physics estimates for the process and orders of magnitude below the previous work. The prospect for practical use of nuclear isomers as energy-storage devices, triggered on demand, is still speculation.

This work was supported in part by the U.S. Department of Energy (DOE) under Contract No. W-31-109-ENG-38 (ANL), W-7405-ENG-48 (UC-LLNL), and W-7405-ENG-36 (UC-LANL), and in part by the DOE Nuclear Energy Research Initiative (NERI).

Principal publication: "Search for X-ray Induced Acceleration of the Decay of the 31-yr Isomer of ^{178}Hf Using Synchrotron Radiation," Phys. Rev. Lett. **87**, 072503 (2001).

References

[1] See, e.g., Science **283**, 769 (1999).

[2] C.B. Collins et al., Phys. Rev. Lett. **82**, 695 (1999).

[3] C.B. Collins et al., Laser Physics **9**, 8 (1999); C.B. Collins et al., Phys. Rev. C **61**, 054305 (2000); C.B. Collins et al., Phys. At. Nucl. **63**, 2067 (2000).

[4] D.P. McNabb et al., Phys. Rev. Lett. **84**, 2542 (2000); P. von Neumann-Cosell and A. Richter, Phys. Rev. Lett. **84**, 2543 (2000).

[5] B. Lai et al., Argonne National Laboratory Report ANL/APS/TB-3 (1993); R.J. Dejus et al., Argonne National Laboratory Report ANL/APS/TB-17 (1994).

[6] I. Ahmad et al., Phys. Rev. Lett. **87**, 072503 (2001).

I. Ahmad,[1] J. C. Banar,[2] J. A. Becker,[3] D. S. Gemmell,[1] A. Kraemer,[3] A. Mashayekhi,[4] D. P. McNabb,[3] G. G. Miller,[2] E. F. Moore,[1] L. N. Pangault,[2] R. S. Rundberg,[2] J. P. Schiffer,[1] S. D. Shastri,[4] T. F. Wang,[3] J. B. Wilhelmy[2]

[1] *Physics Division, Argonne National Laboratory, Argonne, IL, U.S.A.*
[2] *Los Alamos National Laboratory, Los Alamos, NM, U.S.A.*
[3] *Lawrence Livermore National Laboratory, Livermore, CA, U.S.A.*
[4] *Advanced Photon Source, Argonne National Laboratory, Argonne, IL, U.S.A.*